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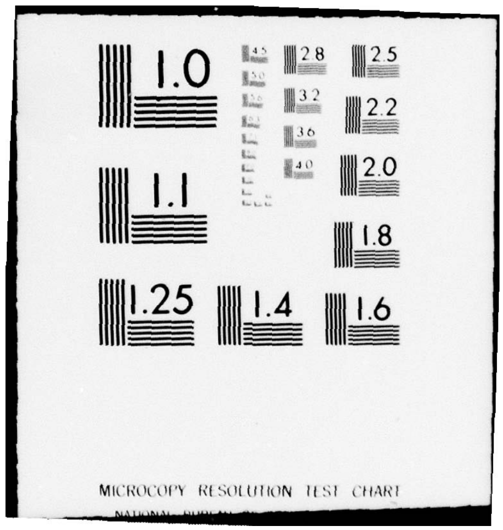
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LEVEL II

PERMEABILITY, MAGNETOMECHANICAL COUPLING AND MAGNETOSTRICTION IN GRAIN-ORIENTED RARE EARTH-IRON ALLOYS

BY H. T. SAVAGE R. ABBUNDI A. E. CLARK

RESEARCH AND TECHNOLOGY DEPARTMENT

1 NOVEMBER 1978

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Prepared for: NAVAL RESEARCH LABORATORY
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Grain-oriented samples of highly magnetostrictive rare earth-iron compounds have successfully been prepared. These samples possess lower inhomogeneous strains than found in the random polycrystalline RFe_2 compounds, resulting in much higher values of relative permeability (μ_r*) and magnetomechanical coupling (k_{33}). A partially oriented $Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$ sample was prepared using a pyrolytic Bridgman type boron nitride crucible. At a bias field of 100 Oe k_{33} = .73, which is considerably

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* mu sub r

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larger than found in the random polycrystal of the same composition. A relative permeability of 36 occurs in this same sample when a low ac drive of 1.6 Oe rms is used. A second fabrication method using a horizontal zone technique with a supporting "cold finger" was employed to grow the ternary $Tb_{.27}Dy_{.73}Fe_{1.98}$. The low ac drive values of k_{33} and μ_r^* were .74 and 19 respectively. The permeability at low bias was found to possess a sharp ac drive dependence. Near zero bias, when the drive was changed from 1.6 Oe rms to 13 Oe rms, μ_r^* in the quaternary increased from 36 to 98. In the ternary the values of μ_r^* near zero bias increased from 19 to 61. Magnetostriction measurements on both samples show a significant increase in $d\lambda/dH$ and λ_s over the random polycrystals.

lambda gamma

* mu sub r

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SUMMARY

The magnetic and magnetomechanical properties study reported here is part of a program to develop magnetostrictive materials for high power sonar projectors. The rare earth iron Laves phase compounds containing terbium possess high magnetostrictions and when they are prepared in textured form become excellent candidates for driving elements of high strain transducers.

This paper reports the permeability, magnetomechanical coupling factor and magnetostriction of compounds prepared by two methods. The coupling factors which are nominally .55-.60 in randomly oriented polycrystals, are increased to .72-.74 in the aligned samples. The initial permeabilities, in like manner, are increased from 6-10 to between 60-100 upon preferential alignment. This increase, coupled with an increase of d-constant to $8\pi \times 10^{-6}$/Oe, will allow a substantial improvement in efficiency and energy density of magnetostrictive drivers.

This study was carried out in the Solid State Branch of the Radiation Division. The materials development was sponsored by the NRL Material Block Program (Howard Lessoff). Magnetic measurements and the fabrication of prototype transducer components were carried out under the sponsorship of the NOSC Transducer Block Program (R. Smith). Research on the magnetoelastic properties of highly magnetostrictive rare earths is sponsored by the Office of Naval Research (B. MacDonald) and the NSWC Independent Research Fund (L. Hill).

Paul R. Wessel

PAUL R. WESSEL
By direction

INTRODUCTION

The cubic Laves phase rare earth-Fe_2 compounds are known to possess large room temperature magnetostriction constants (λ) as well as huge values of magnetic anisotropy (K).[1] However with an appropriate choice of compounds, ternaries of the form $R_xR_{1-x}Fe_2$ may be obtained which retain the large values of λ but possess anisotropies which are two orders of magnitude lower than in the RFe_2 compounds.[1] Similarly, the addition of another rare earth $R_xR_yR_{1-x-y}Fe_2$ further lowers the anisotropy.[2] These ternary and quaternary compounds are characterized by high values of magnetomechanical coupling, k_{33}.[3,4] The magnetostriction is quite anisotropic with $\lambda_{100} \approx \lambda_{111}/10$. In the ternary to be reported on here $\lambda_{111} = 1.6 \times 10^{-3}$ and the quaternary $\lambda_{111} = .8 \times 10^{-3}$. However the large magnetostriction in the random polycrystals of these materials results in a rather low value of relative permeability.[3,4]

In this paper we report on two fabrication methods which have been successful in preparing rare earth-iron compounds with a large degree of grain orientation. These samples thus possess much lower inhomogeneous strains that are found in the corresponding random polycrystals, resulting in much higher values of μ_r and k_{33}. They also exhibit a significant increase in $d\lambda/dH$ as well as in λ_s.

1. Clark, A. E., "Magnetic and Magnetoelastic Properties of Highly Magnetostrictive Rare Earth-Iron Laves Phase Compounds," AIP Conf. Proc., Vol 18, 1974, p. 1015.

2. Williams, C. M. and Koon, N. C., "Anisotropy of Single Crystal $Ho_xDy_yTb_{1-x-y}Fe_2$ Laves Phase Compounds," Physica, Vol. 86, 1977, p. 147.

3. Savage, H. T., Clark, A. E., and Powers, J. M., "Magnetomechanical Coupling and ΔE Effect in Highly Magnetostrictive Rare Earth-Fe_2 Compounds," IEEE Trans. on Magnetics, Vol. MAG-11, 1975, p. 1355.

4. Savage, H. T., Clark, A. E., Koon, N. C., and Williams, C. M., "The Temperature and Composition Dependence of the Magnetomechanical Coupling Factor in Rare Earth-Fe_2 Alloys," IEEE Trans. on Magnetics, Vol. MAG-13, 1977, p. 1517.

SAMPLE FABRICATION

In our first method a partially grain-oriented $Tb_{.27}Dy_{.73}Fe_{1.98}$ sample was prepared by a horizontal zone method, using a supporting "cold finger" to hold the zone in place. The zone was passed through the originally arc-cast material at the rate of about 1 cm/minute. The resulting boule was elliptical in shape, approximately 10 cm long and 0.6 cm in average diameter. The sample possessed a grain structure with a strong preferential orientation. The grains are not equiaxed but elongated with an aspect ratio of from 2-1 to 5-1. The long axis varied roughly from 0.5 to 2 mm. The direction of the long axis of the grains in the top half of the boule lie at small angles relative to the boule axis. However, a substantial change in the grain orientation was found to occur in the half of the boule nearest the "cold finger" influences grain orientation.

A second method of preparation, Bridgman in nature, was used to prepare a sample of $Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$. A boron nitride crucible containing the melt was dropped through a temperature gradient at a rate of .2 cm/min. The resultant boules were from 5 to 8 cm long with an average cross section of about 1 cm. The boule is almost single crystal in nature with small angle grain boundaries of less than 5°. A <111> direction was found perpendicular to the growth axis. The growth axis is about <123>. Coupling factor measurements (to be discussed later) show this to be a favorable growth axis. This method of preparation yields a large relatively homogeneous boule that could be used in its entirety. The horizontal zone method does not yield a homogeneous boule.

A reoccurring problem has been the appearance of a Widmanstatten precipitate (WSP) which we believe to be a rare-earth Fe_3 compound. In mixing a series of alloys of rare-earth Fe_{2-x} we find no WSP for $x > .05$. WSP was always present for $x < .05$ when the method of growth for the series was a modification of the Bridgman technique. The starting mixture for the horizontal zone was $x = .02$. No WSP was found. The zone was moved rapidly enough that (perhaps) less rare earth was lost with this technique. The chemical formulae given in the text are the starting mixtures.

EXPERIMENTAL RESULTS

The $Tb_{.27}Dy_{.73}Fe_{1.98}$ sample was cut horizontally along the boule axis so that measurements could be made on the top half, in which the grains point along the axis. The peak relative permeability of this section was found to be 19 when a low ac drive of 1.6 Oe rms was used. However a substantial increase in μ_r was seen as the ac drive was increased. Figure 1 shows the relative permeability at constant stress as a function of the applied bias field for a 1.6 and 13 Oe rms ac drive level. Using the higher drive results in a $\mu_r = 61$, with the peak occurring at a very low bias field of ≈ 2 Oe. These values represent at least a 2-fold increase in relative permeability over the random polycrystal of the same composition in which $\mu_r = 6$ to 10.[4] Quite similar results were obtained for the $Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$ compound as shown in Fig. 2, where μ_r is again plotted for two different drive levels. At the low ac drive of 1.6 Oe rms the relative permeability of 98 was obtained with a 13 Oe rms ac drive. This sharp increase only occurs for a bias field < 100 Oe. At higher bias μ_r shows little ac drive dependence. This behavior is true for both the ternary as well as the quaternary and we believe that this is the first time that such a dramatic increase in μ_r (as a function of drive) has been observed. We speculate that internal strains are being overcome, allowing the domain walls to move easily in comparison with the low drive situation.

Both of these partially oriented samples were found to possess substantially larger values of magnetomechanical coupling (k_{33}) than has previously been observed in their random polycrystalline counterparts.[3,4] Figure 3 shows a comparison of k_{33}^2 as a function of applied bias between these two partially oriented samples and a typical arc-cast $Tb_{.27}Dy_{.73}Fe_2$ random polycrystal. The peak coupling squared in the random polycrystal is $k_{33}^2 = .28$. However in the two oriented samples $k_{33}^2 = .54$. at 125 Oe bias for the ternary, while $k_{33}^2 = .53$ at 100 Oe bias for the quaternary. All the coupling measurements were performed using a 1.6 Oe rms ac drive. The lower part of the boule which had a different grain configuration had a peak value of k_{33}^2 of .43. Apparently the presence of the "cold finger" is detrimental in obtaining optimum grain orientation. The quaternary sample peaks at a slightly smaller bias due to the smaller magnetocrystalline anisotropy for this composition.[2] It should be noted that the peak in the coupling coefficient occurs at a much higher bias than the peak relative permeability. This is brought about by the fact that the coupling is essentially zero when the magnetic moment is zero.

As previously stated the composition for both alloys were chosen to minimize the anisotropy yet maintain the large room temperature magnetostriction. Figure 4 shows the results of magnetostriction measurements on the partially oriented $Tb_{.27}Dy_{.73}Fe_{1.98}$ sample. The strain as a function of applied field is shown for two different strain gauge locations positioned along the axis of the sample. The upper curves show $\lambda_{||}$, which is the strain obtained when the applied field is

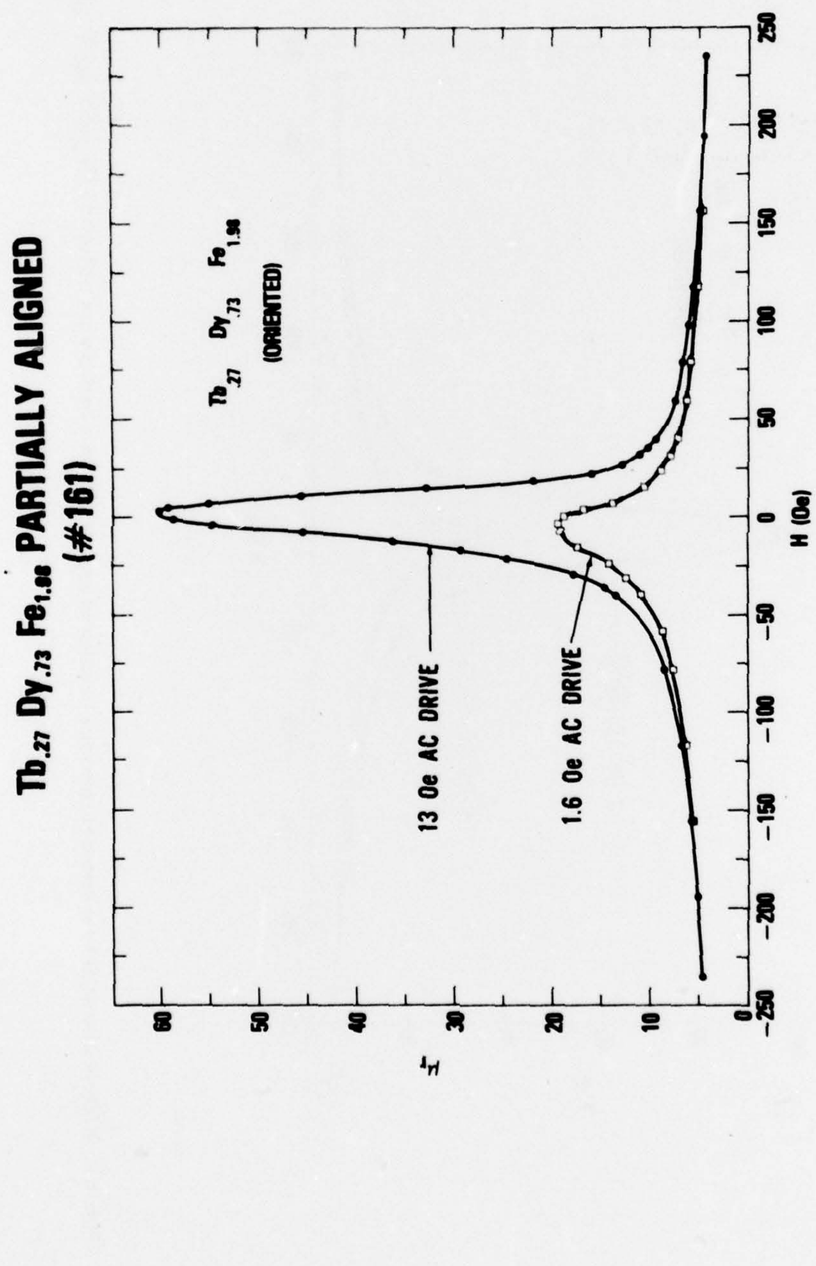

Figure 1. Relative permeability at constant stress as a function of applied bias for a partially grain-oriented $Tb_{.27}Dy_{.73}Fe_{1.98}$ sample.

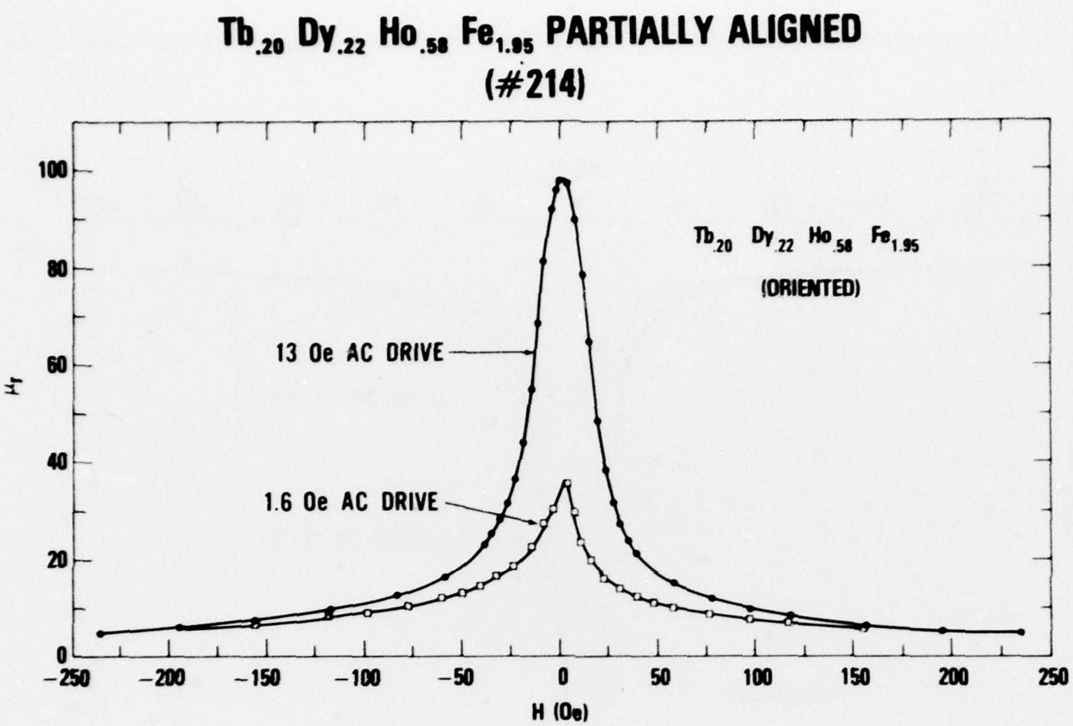

Figure 2. Relative permeability at constant stress as a function of applied bias for a partially grain-oriented $Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$ sample.

directed along the axis of the rod. The lower curves, $\lambda_\perp$, are the strains obtained when the field is applied perpendicular to the long axis of the sample. As can be seen in the figure, the large demagnetizing field $\approx$ 4.5 kOe that results when the field is applied perpendicular to the long axis of the rod, prevents saturation value of $\lambda_\perp$ to be -800×10^{-6} for both grains. Figure 4 clearly indicates that the gauges sampled grains with quite different orientations. Location #1 was obviously a very favorable grain orientation yeilding $\lambda_s \equiv \frac{2}{3}(\lambda_{||} - \lambda_\perp) = 1.3 \times 10^{-3}$. This value of λ_s is equal to .83 λ_{111}, indicating that the orientation is close to a <111> direction. Less favorable results were obtained for location #2 where $\lambda_s = 1.1 \times 10^{-3}$. Measurements on random polycrystals yeild a $\lambda_s = 1.0 \times 10^{-3}$.

The d constant is defined as the slope of the magnetostriction curve ($4\pi\, d\lambda/dH$). The d constant is an important figure of merit in applications. In the vicinity of maximum coupling values of d are somewhat greater than $8\pi \times 10^{-6}$ Oe^{-1} in location 1 and somewhat greater than 10^{-6} Oe^{-1} in region 2. Figure 5 shows a plot of the magnetostriction as a function of applied field for the oriented $Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$ sample. Both $\lambda_{||}$ and $\lambda_\perp$ are easily saturated with available fields due to the small magnetic anisotropy this composition possesses. The saturation magnetostriction $\lambda_s = .74 \times 10^{-3}$ represents $.9\lambda_{111}$ and demonstrates the high degree of orientation in this sample. This value of λ_s is within 5% of the value of λ_s calculated from our x-ray determination of the growth axis. Only one location in this sample was investigated due to the large and regular grain structure. The d constant was found to be $6\pi \times 10^{-6}$ Oe^{-1}. These values of the d constant are to be compared with values of somewhat less than $4\pi \times 10^{-6}$ Oe^{-1} in random polycrystals.

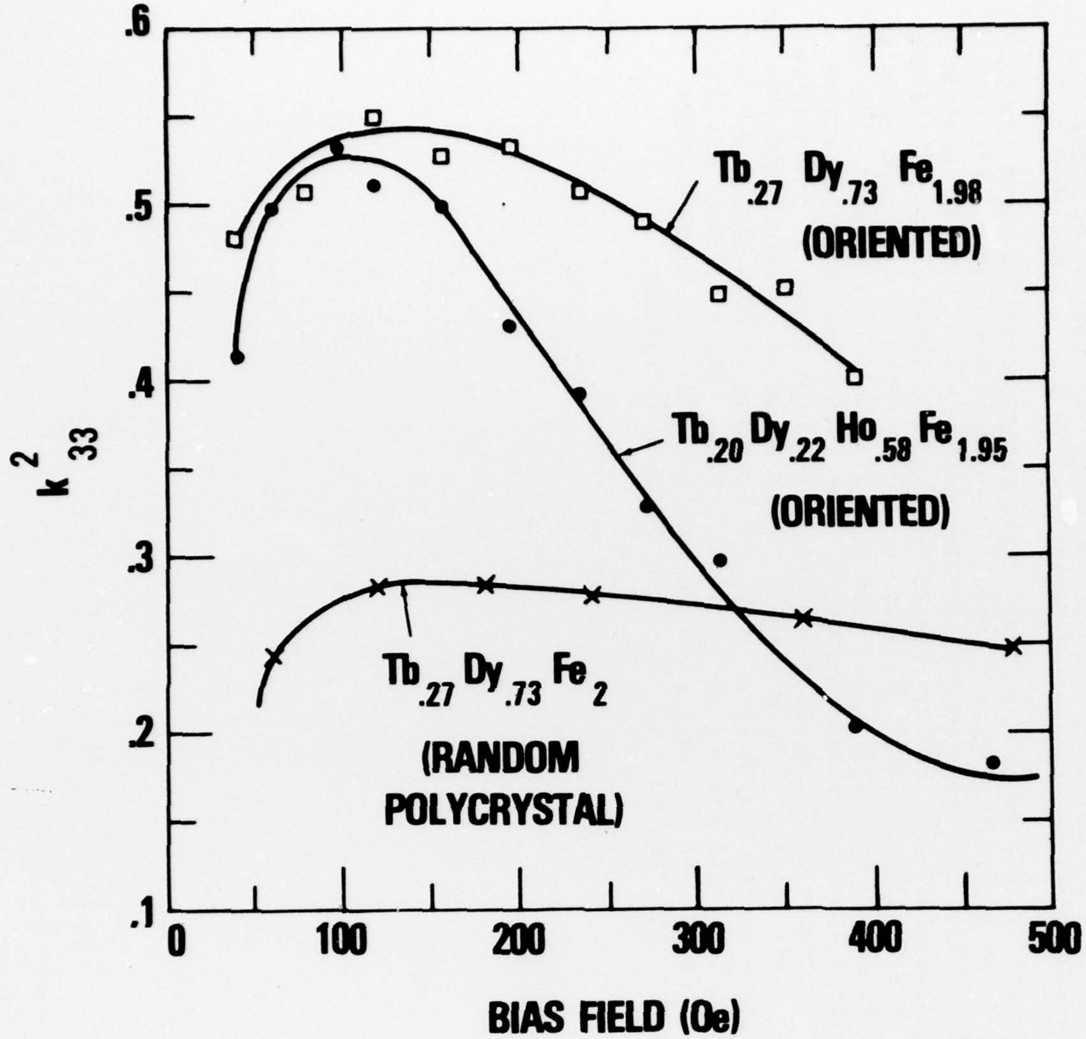

Figure 3. A comparison of the magneto-mechanical coupling factor k^2_{33} as a function of applied bias between the partially grain-oriented $Tb_{.27}Dy_{.73}Fe_{1.98}$ and $Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$ samples and a $Tb_{.27}Dy_{.73}Fe_2$ random polycrystal.

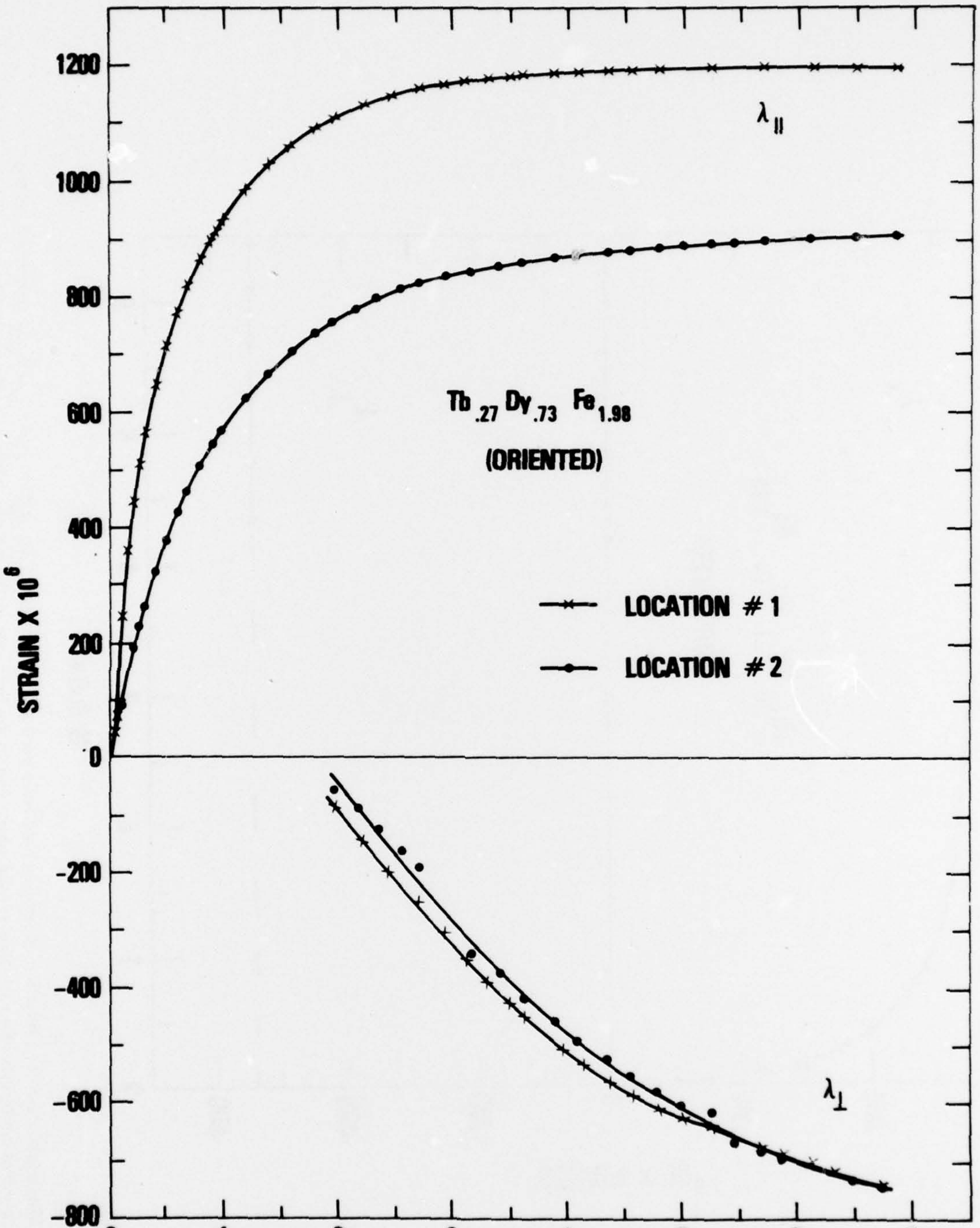

Figure 4. Magnetostriction as a function of applied field for a partially grain-oriented $Tb_{.27}Dy_{.73}Fe_{1.98}$ sample. Strain gauges were positioned at two different locations along the sample. $\lambda_{\parallel}$ is the strain which results when the field is applied parallel to the long axis of the sample, while $\lambda_{\perp}$ is the strain when the field is applied perpendicular to the long axis.

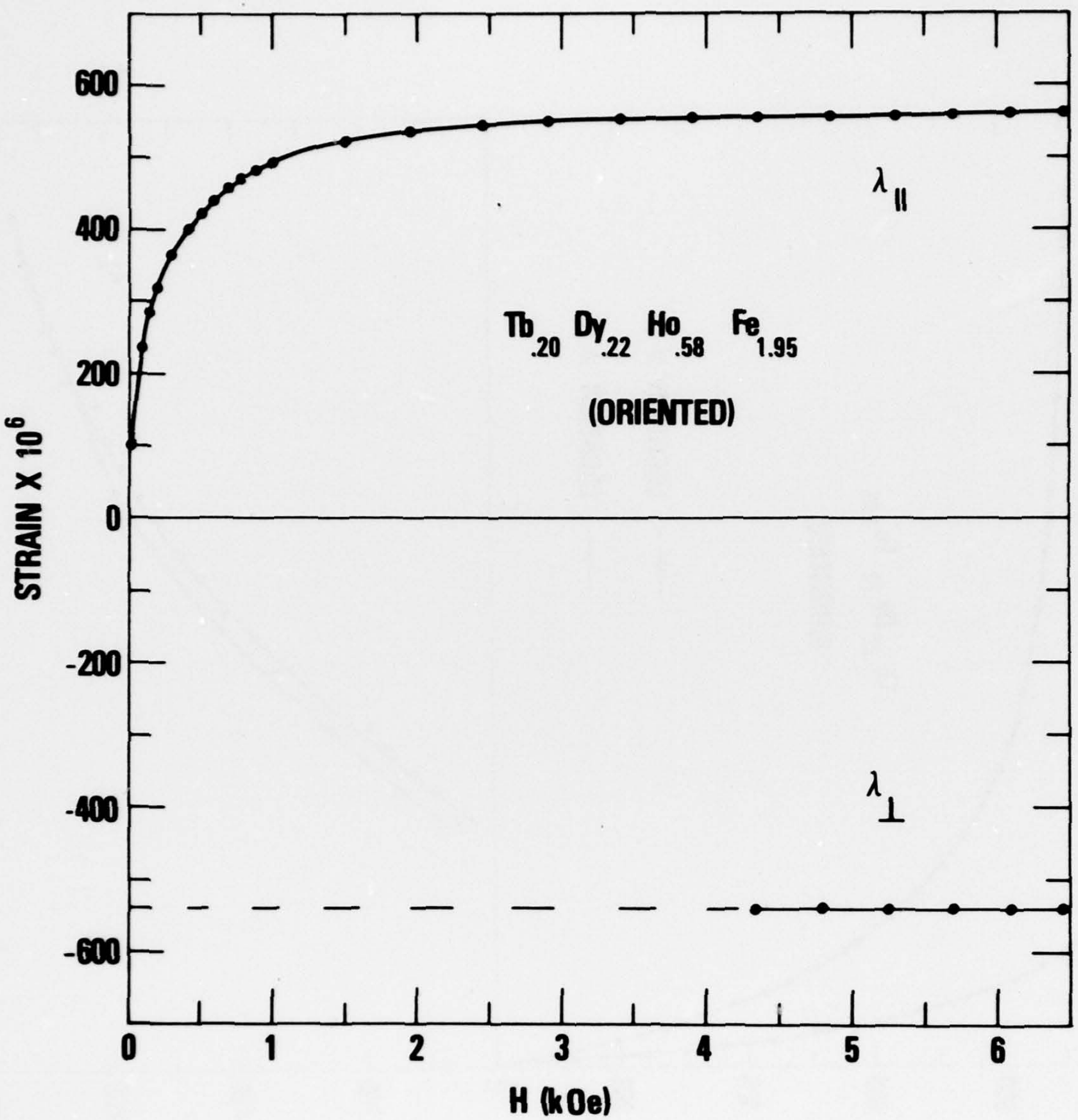

Figure 5. Magnetostriction as a function of applied field for a partially grain-oriented $Tb_{.20}Dy_{.22}Ho_{.58}Fe_{1.95}$ sample. $\lambda_{\parallel}$ is the strain which results when the field is applied parallel to the long axis of the sample, while $\lambda_{\perp}$ is the strain when the field is applied perpendicular to the long axis.

RECOMMENDATIONS

A magnetostriction of 1000 ppm at 500 Oe requires a d constant of $8\pi \times 10^{-6}$/Oe. A sample of Terfenol-D achieving this performance would have an energy density (including copper drive coil) of 3700 J/m^3 with a 20 kA/m bias field and 12 kA/m rms drive field.[5] This is a five-fold increase over that currently available from PZT-4[5] (670 J/m^3) operating at drive conditions of comparable difficuly (0.5 MV/m rms).[5] *It is recommended that the materials program aim at developing* RFe_2 alloys possessing this value of d constant.

In the quaternary sample reported here, $d = 6\pi \times 10^{-6}$/Oe; for the ternary sample $4\pi < d < 8\pi$ ($\times 10^{-6}$/Oe). The goal of $d = 8\pi \times 10^{-6}$/Oe appears attainable with refinement of the techniques discussed above. Upon achieving this goal, the problem of brittleness should be addressed. While there was no breakage of rare earth elements during construction of the Raytheon prototype 50 - 100 W transducer, less care would be required to handle more rugged magnetostrictive elements.[6]

5. Meeks, S., "Rare Earth Iron Materials," USRD/NRL Memorandum to S02-38 File, 13 Feb 1978.

6. Butler, J., and Ciosik, S., "Rare Earth Magnetostrictive Transducer," Final Report, NOSC Contract N66001-77-C-0095, Oct 1978.

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